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# A short note on spin pumping theory with Landau-Lifshitz-Gilbert equation under quantum fluctuation; necessity for quantization of localized spin

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## Abstract

We would like to point out the blind spots of the approach combining the spin pumping theory proposed by Tserkovnyak et al. with the Landau-Lifshitz-Gilbert equation; this method has been widely used for interpreting vast experimental results. The essence of the spin pumping effect is the quantum fluctuation. Thus, localized spin degrees of freedom should be quantized, i.e. be treated as magnons not as classical variables. Consequently, the precessing ferromagnet can be regarded as a magnon battery. This point of view will be useful for further progress of spintronics.

## 1 Introduction

A standard way to generate a spin current is the spin pumping effect at the interface between a ferromagnet and a non-magnetic material. The precessing ferromagnet acts as a source of spin angular momentum to induce a spin current pumped into a non-magnetic material, under the ferromagnetic resonance; *spin battery*. [1] This method was first theoretically proposed by Silsbee et al. [2] and have been developed by Tserkovnyak et al. Though Tserkovnyak et al. have phenomenologically treated the spin-flip scattering processes, [3] now their spin pumping theory has been widely used for interpreting vast experimental results, [4, 5, 6, 7] in particular by experimentalists. Thus it will be useful to point out the blind spots of their phenomenological [6] formula under time-

dependent transverse magnetic fields (i.e. quantum fluctuations), for further progress of spintronics. This is the main purpose of this paper.

This manuscript is structured as follows. We point out the blind spots of the approach combining the spin pumping theory proposed by Tserkovnyak et al. with the Landau-Lifshitz-Gilbert (LLG) equation in sec. 2. We show that, on the basis of the Schwinger-Keldysh formalism, the essence of the spin pumping effect is quantum fluctuations in sec. 3. We also discuss why the approach combining the theory by Tserkovnyak et al. with the LLG eq. cannot appropriately describe the spin pumping effect and propose an alternative way to capture the dynamics.

In this paper, we use the term, *quantum fluctuations*, to indicate time-dependent transverse magnetic fields. In addition, we regard the z-axis as the quantization axis, and take  $\hbar = 1$ .

## 2 Spin Pumping Theory with Landau-Lifshitz-Gilbert Equation

According to the phenomenological[6] spin pumping theory by Tserkovnyak et al. and their notation,[8, 3] the pumped spin current  $\mathbf{I}_{\text{s-pump}}$  reads

$$\mathbf{I}_{\text{s-pump}} = G_{\perp}^{(\text{R})} \mathbf{m} \times \dot{\mathbf{m}} + G_{\perp}^{(\text{I})} \dot{\mathbf{m}}, \quad (1)$$

where the dot denotes the time derivative. We have taken  $e = 1$ , and  $\mathbf{m}(\mathbf{x}, t)$  denotes a unit vector along the magnetization direction; they have treated  $\mathbf{m}(\mathbf{x}, t)$  as classical variables. The variable  $G_{\perp}$  is the complex-valued mixing conductance that depends on the material;[9, 10]  $G_{\perp} = G_{\perp}^{(\text{R})} + iG_{\perp}^{(\text{I})}$ . In addition, they suppose that the magnetization dynamics of ferromagnets can be described by the LLG eq.;

$$\dot{\mathbf{m}} = \gamma \mathbf{H}_{\text{eff}} \times \mathbf{m} + \alpha \mathbf{m} \times \dot{\mathbf{m}}, \quad (2)$$

where  $\gamma$  is the gyro-magnetic ratio and  $\alpha$  is the Gilbert damping constant that determines the magnetization dissipation rate.

Here it should be emphasized that though this Gilbert damping constant,  $\alpha$ , was phenomenologically introduced,[11] it can be derived microscopically by considering a whole system including spin relaxation;[12] thus the effect of the exchange coupling to conduction electrons should be considered to have already been included into this Gilbert damping term. In addition, Rebei et al.[13] have started from a quantum model and have shown that the Gilbert damping term arises only in the limit of small deviations from local equilibrium, where fluctuations are negligible. In other words, only in the classical limit,[14] the LLG eq., eq. (2), is an appropriate way to describe the dynamics of the ferromagnet.

The effective magnetic field is set as

$$\mathbf{H}_{\text{eff}} = (\Gamma(t), 0, B), \quad (3)$$

where  $\Gamma(t)$  represents a time-dependent transverse magnetic field. The LLG eq. becomes

$$\begin{pmatrix} \dot{m}^x \\ \dot{m}^y \\ \dot{m}^z \end{pmatrix} = \gamma \begin{pmatrix} -Bm^y \\ Bm^x - \Gamma m^z \\ \Gamma m^y \end{pmatrix} + \alpha \begin{pmatrix} m^y \dot{m}^z - \dot{m}^y m^z \\ m^z \dot{m}^x - \dot{m}^z m^x \\ m^x \dot{m}^y - \dot{m}^x m^y \end{pmatrix}. \quad (4)$$

Eq. (4) is substituted into  $I_{s\text{-pump}}^z$ , eq. (1); we include the contribution of the Gilbert damping term, which depends on the materials, up to  $\mathcal{O}(\alpha)$ ;  $\alpha \sim 10^{-3}, 10^{-2}$  for  $\text{Ni}_{81}\text{Fe}_{19}$  (metal), [4] and  $\alpha \sim 10^{-5}$  for  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  (insulator), [15] as examples. Their theory is applicable to both ferromagnetic metals and insulators. [1]

Then the z-component of each term in eq. (1) becomes

$$\begin{aligned} (\mathbf{m} \times \dot{\mathbf{m}})^z &= \gamma B[(m^x)^2 + (m^y)^2] - \gamma \Gamma m^x m^z - \alpha \gamma \Gamma [m^x m^y m^x + (m^y)^3 + (m^z)^2 m^y] + \mathcal{O}(\alpha^2) \\ \dot{m}^z &= \gamma \Gamma m^y + \alpha \gamma \{B[(m^x)^2 + (m^y)^2] - \Gamma m^x m^z\} + \mathcal{O}(\alpha^2). \end{aligned} \quad (6)$$

Consequently, the z-component of the pumped spin current reads

$$\begin{aligned} I_{s\text{-pump}}^z &= G_{\perp}^{(R)} \left\{ \gamma B[(m^x)^2 + (m^y)^2] - \gamma \Gamma m^x m^z - \alpha \gamma \Gamma [m^x m^y m^x + (m^y)^3 + (m^z)^2 m^y] \right\} \\ &+ G_{\perp}^{(I)} \left\{ \alpha \gamma \{B[(m^x)^2 + (m^y)^2] - \Gamma m^x m^z\} + \gamma \Gamma m^y \right\} + \mathcal{O}(\alpha^2). \end{aligned} \quad (7)$$

$$\xrightarrow{\Gamma \rightarrow 0} [G_{\perp}^{(R)} + \alpha G_{\perp}^{(I)}] \gamma B[(m^x)^2 + (m^y)^2]. \quad (8)$$

At finite temperature, the magnetization is thermally activated;  $\dot{\mathbf{m}} \neq 0$ . [16] Then the time derivative of the z-component, eq. (6), means

$$\dot{m}^z = \gamma \Gamma m^y + \alpha \gamma \{B[(m^x)^2 + (m^y)^2] - \Gamma m^x m^z\} + \mathcal{O}(\alpha^2). \quad (9)$$

$$\xrightarrow{\Gamma \rightarrow 0} \alpha \gamma B[(m^x)^2 + (m^y)^2]. \quad (10)$$

$$\neq 0. \quad (11)$$

Eqs. (8), (10), and (11) mean that, within the framework by Tserkovnyak et al. with the LLG eq., they may gain spin currents at finite temperature if only the magnetic field along the z-axis,  $B$ , is applied;

$$I_{s\text{-pump}}^z \xrightarrow{\Gamma \rightarrow 0 (B \neq 0)} 0. \quad (12)$$

That is, the approach combining the spin pumping theory by Tserkovnyak et al. with the LLG eq. concludes that spin currents may be pumped at finite temperature without time-dependent transverse magnetic fields.

## 3 Quantum Spin Pumping Mediated by Magnon

### 3.1 Spin pumping via the Schwinger-Keldysh formalism

On the other hand, our spin pumping theory [17] mediated by magnons denies the possibility. We have considered a ferromagnetic insulator and non-magnetic

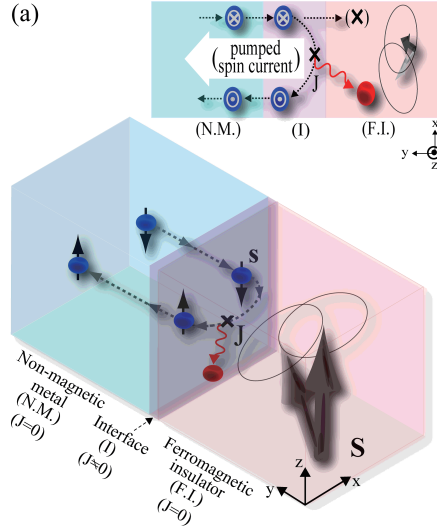


Figure 1: (Color online) A schematic picture of the spin pumping effect mediated by magnons (a), and the inverse process (b). Spheres represent magnons and those with arrows are conduction electrons. The interface is defined as an effective area where the Fermi gas (conduction electrons) and the Bose gas (magnons) coexist to interact;  $J \neq 0$ . Conduction electrons cannot enter the ferromagnet, which is an insulator.

metal junction;

$$\mathcal{H}_{\text{ex}} = -2Ja_0^3 \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} \mathbf{S}(\mathbf{x}, t) \cdot \mathbf{s}(\mathbf{x}, t), \quad (13)$$

where  $2J$  represents the exchange interaction between localized spins ( $\mathbf{S}(\mathbf{x}, t)$ ,  $\mathbf{x} = (x, y, z) \in \mathbf{R}^3$ ) and conduction electrons ( $\mathbf{s}$ ), and  $a_0$  denotes the lattice constant among ferromagnets (see Fig. 1). Conduction electron spin variables are represented as

$$s^j = \sum_{\eta, \zeta = \uparrow, \downarrow} [c_{\eta}^{\dagger}(\sigma^j)_{\eta\zeta} c_{\zeta}] / 2 \quad (14)$$

$$\equiv (c^{\dagger} \sigma^j c) / 2, \quad (15)$$

where  $\sigma^j$  are the  $2 \times 2$  Pauli matrices;

$$[\sigma^j, \sigma^k] = 2i\epsilon_{jkl}\sigma^l, (j, k, l = x, y, z). \quad (16)$$

Operators  $c^{\dagger}/c$  are creation/annihilation operators for conduction electrons, which satisfy the (fermionic) anticommutation relation;

$$\{c_{\eta}(\mathbf{x}, t), c_{\zeta}^{\dagger}(\mathbf{x}', t)\} = \delta_{\eta, \zeta} \delta(\mathbf{x} - \mathbf{x}'). \quad (17)$$

We suppose the uniform magnetization and thus localized spin degrees of freedom can be mapped into magnon ones via the Holstein-Primakoff transformation;

We have microscopically calculated the pumped spin current mediated by magnons on the basis of the Schwinger-Keldysh formalism[18, 19, 20, 21, 22, 23] with a time dependent transverse magnetic field;

$$\Gamma(t) = \Gamma_0 \cos(\Omega t), \quad (18)$$

which acts as a quantum fluctuation. Our spin pumping theory at finite temperature gives[17] (the concrete calculation has been shown in our previous work[17] in detail, and please see it)

$$\mathcal{T}_s^z \xrightarrow{\Gamma \rightarrow 0(B \neq 0)} 0 + \mathcal{O}(J^2), \quad (19)$$

where  $\mathcal{T}_s$  represents the spin transfer torque; by integrating over the interface, the pumped spin current can be estimated.[24, 17] Thus our formalism shows that even when the magnetic field along the quantization axis (z-axis),  $B$ , is applied, spin currents mediated by magnons cannot flow without quantum fluctuations (i.e. time-dependent transverse magnetic fields). This is the main difference from the theory by Tserkovnyak et al. with the LLG eq., eq. (12). Quantum fluctuations induce (net) spin currents, and are essential to spin pumping mediated by magnons as well as the exchange interaction;[17]

$$\mathcal{T}_s^z \propto J\Gamma_0^2 \quad (20)$$

$$\xrightarrow{\Gamma \rightarrow 0(B \neq 0)} 0. \quad (21)$$

Therefore we call our spin pumping theory *quantum spin pumping*. [17]

Moreover, it will be useful to mention that we have revealed that  $\mathcal{T}_s^z$  has a resonance structure as a function of the angular frequency of the applied transverse field; the resonance condition reads,  $\Omega = J$ , because the localized spin acts as an effective magnetic field along the quantization axis,  $J$ , for conduction electrons. This fact (i.e. resonance condition) is useful to enhance the quantum spin pumping effect because the angular frequency of a transverse magnetic field is under our control.

In addition, the work by Adachi et al.[25, 26] supports our result; they have adopted the same approach with ours[17] (i.e. the Schwinger-Keldysh formalism with magnon degrees of freedom) and have studied the contribution of the exchange interaction,  $J$ , to spin pumping up to  $\mathcal{O}(J^2)$  without time-dependent transverse magnetic fields (i.e. quantum fluctuations). They also suppose the uniform magnetization and thus localized spin degrees of freedom can be mapped into magnon ones via the Holstein-Primakoff transformation;

$$S^+(\mathbf{x}, t) \equiv S^x(\mathbf{x}, t) + iS^y(\mathbf{x}, t) \quad (22)$$

$$= \sqrt{2\tilde{S}}a(\mathbf{x}, t) + \mathcal{O}(\tilde{S}^{-1/2}), \quad (23)$$

$$S^-(\mathbf{x}, t) \equiv S^x(\mathbf{x}, t) + iS^y(\mathbf{x}, t) \quad (24)$$

$$= \sqrt{2\tilde{S}}a^\dagger(\mathbf{x}, t) + \mathcal{O}(\tilde{S}^{-1/2}), \quad (25)$$

$$S^z(\mathbf{x}, t) = \tilde{S} - a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t), \quad (26)$$

$\tilde{S} \equiv S/a_0^3$ , where operators  $a^\dagger/a$  are magnon creation/annihilation operators satisfying the (bosonic) commutation relation;

$$[a(\mathbf{x}, t), a^\dagger(\mathbf{x}', t)] = \delta(\mathbf{x} - \mathbf{x}'). \quad (27)$$

Up to the  $\mathcal{O}(S)$  terms, localized spins reduce to a free boson system.

As the result, the exchange interaction between localized spins and conduction electrons,  $\mathcal{H}_{\text{ex}} (\equiv \mathcal{H}_{\text{ex}}^S + \mathcal{H}_{\text{ex}}')$ , is rewritten as

$$\mathcal{H}_{\text{ex}}^S = -JS \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} c^\dagger(\mathbf{x}, t) \sigma^z c(\mathbf{x}, t), \quad (28)$$

$$\mathcal{H}_{\text{ex}}' = -Ja_0^3 \sqrt{\frac{\tilde{S}}{2}} \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} [a^\dagger(\mathbf{x}, t)c^\dagger(\mathbf{x}, t)\sigma^+ c(\mathbf{x}, t) + a(\mathbf{x}, t)c^\dagger(\mathbf{x}, t)\sigma^- c(\mathbf{x}, t)]. \quad (29)$$

This,  $\mathcal{H}_{\text{ex}}'$ , means that localized spins lose spin angular momentum by emitting magnons and conduction electrons flip from down to up by absorbing the momentum (Fig. 1 (a)), and vice versa (Fig. 1 (b)).

They[25, 26] have revealed, through the standard procedure of the Schwinger-Keldysh formalism, that spin currents cannot be generated under the thermal equilibrium condition where the temperature difference does not exist between localized spins and conduction electrons, because of the balance between thermal fluctuations in ferromagnet and those in non-magnetic metal, even when a magnetic field along the quantization axis is applied;[25, 26, 17]

$$\mathcal{T}_s^z \stackrel{B \neq 0}{=} 0 + \mathcal{O}(J^4). \quad (30)$$

That is, a net spin current is induced by inhomogeneous thermal fluctuations between conduction electrons and magnons, not by the the applied magnetic field along the quantization axis  $B$ , also in this case.

### 3.2 Necessity for the quantization of localized spins; magnon

Though the approach combining the spin pumping theory proposed by Tserkovnyak et al. with the LLG eq. has insisted that spin currents may be pumped at finite temperature if only the magnetic field along the z-axis,  $B$ , is applied (see eq. (12)), our spin pumping theory mediated by magnons[17, 25, 26] has denied the possibility (see eqs. (19) and (30)). Here it should be emphasized that the Schwinger-Keldysh formalism,[18, 19, 20, 21, 22] which we have adopted,[17, 25, 26] has microscopically captured the (nonequilibrium) spin-flip

dynamics,  $\mathcal{H}'_{\text{ex}}$ , on the basis of the rigorous quantum mechanics beyond phenomenology (see also Fig. 1). In addition as pointed out by Rebei et al.,[13] though the LLG eq. cannot capture the true transient behavior of the system, the Schwinger-Keldysh formalism has microscopically described the time development of the spin pumping effect.[17, 27]

Therefore this result, eqs. (12), (19), and (30), means that the approach combining the spin pumping theory proposed by Tserkovnyak et al. with the LLG eq.[13] is unsuited to the description of the dynamics of the spin pumping effect; due to the phenomenological treatment of the spin-flip scattering processes[3] and the classical treatment of localized spin degrees of freedom.

Spin currents are induced by quantum fluctuations. Then the LLG eq., which treats localized spins as classical variables not as quantized spinwaves (i.e. magnons), cannot capture the dynamics appropriately. Of course  $\mathcal{T}_s^z$  operates the coherent magnon state (as discussed in sec. 3.3), where the uncertainties of the coordinate and of the momentum is a minimum,[28, 29] still, the coherent magnon state has been the quantum state.[30, 13, 14] Thus unless localized spins are quantized, i.e. are treated as magnons,[27] the effect of quantum fluctuations cannot be included correctly.

Here it should be noted that as pointed out in sec. 2, the LLG eq., eq. (2), is an appropriate way to describe the dynamics of the ferromagnet only in the classical limit.[13, 14] Therefore, our approach (i.e. adopting the Schwinger-Keldysh formalism with magnon degrees of freedom) is one of the most valid one to describe the spin pumping effect under time-dependent transverse magnetic fields.

### 3.3 Time evolution of the coherent magnon state

Last, it will be useful to mention that the explicit form[25, 26] of  $\mathcal{T}_s^z$  under the Hamiltonian,  $\mathcal{H}_{\text{ex}} (= \mathcal{H}_{\text{ex}}^S + \mathcal{H}'_{\text{ex}})$ , reads

$$\mathcal{T}_s^z = iJa_0^3 \sqrt{\frac{\tilde{S}}{2}} \langle a^\dagger(\mathbf{x}, t) c^\dagger(\mathbf{x}, t) \sigma^+ c(\mathbf{x}, t) - a(\mathbf{x}, t) c^\dagger(\mathbf{x}, t) \sigma^- c(\mathbf{x}, t) \rangle, \quad (31)$$

where  $\langle \cdots \rangle$  denotes the expectation value estimated for the total Hamiltonian. It is clear that  $\mathcal{T}_s^z$  operates the coherent magnon state. According to Glauber et al.,[31, 32, 33] if the time derivative of the annihilation operator does not involve a functional dependence on the creation operator, i.e., if

$$\dot{a}(t) = f(a(t), t), \quad (32)$$

then the states which are initially coherent remain coherent at all times; that is the coherent state is stable under the time evolution.

Under the Hamiltonian  $\mathcal{H}_{\text{ex}}$  (i.e. eqs. (28) and (29)), the Heisenberg equation of motion reads

$$\dot{a}(\mathbf{x}, t) = iJa_0^3 \sqrt{\frac{\tilde{S}}{2}} c^\dagger(\mathbf{x}, t) \sigma^+ c(\mathbf{x}, t). \quad (33)$$



This means that the condition proposed by Glauber et al. is satisfied; that is the coherent state is stable under the time development.

In addition, also in the case of the quantum spin pumping under time-dependent transverse magnetic fields,[17]  $\mathcal{T}_s^z$  operates the coherent magnon state to have been stable under the time evolution because it satisfies the generalized condition proposed by Mista.[34]

## 4 Conclusion

The essence of the spin pumping effect under time-dependent transverse magnetic fields is not the applied magnetic field along the quantization axis (z-axis) but the quantum fluctuation. As the result, localized spin degrees of freedom should be quantized, i.e. be treated as magnons not as classical variables. From this viewpoint, the precessing ferromagnet can be regarded as *magnon battery*. We consider that the Schwinger-Keldysh formalism is one of the most valid tools to capture the dynamics appropriately.

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